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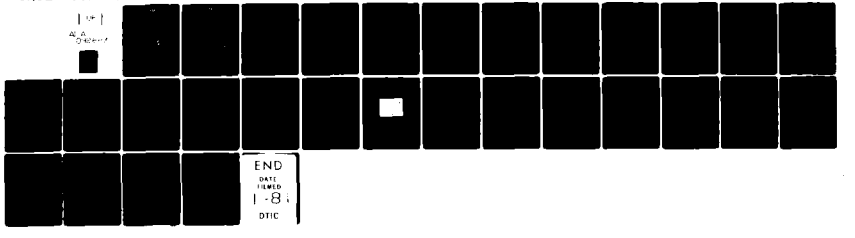

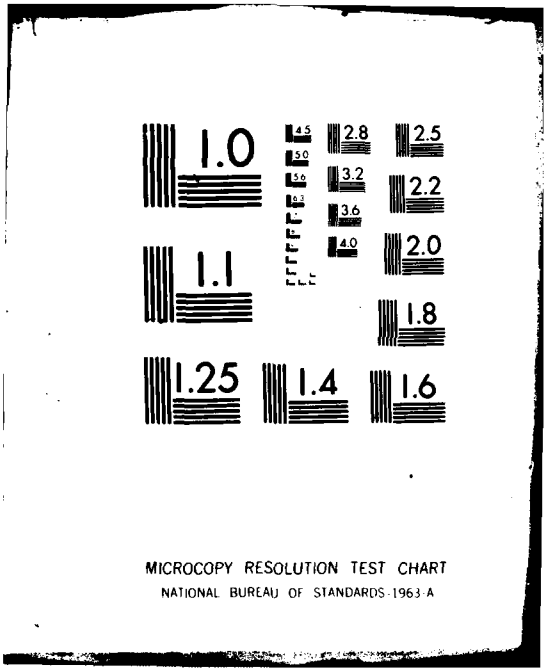
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line broadening, line shifts, recombination, associative excitation, dissociative excitation, Penning ionization, photoionization, metastable interactions.

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

A. A nitrogen-pumped dye laser system has been used to investigate the $3^2P \rightarrow n^2D_{3/2,5/2}$ and $3^2P_{1/2,3/2} \rightarrow n^2S_{1/2}$ ($6 \leq n \leq 22$) line profiles for sodium perturbed by .2 to 2 Amagats of argon. Sodium was ionized in a stepwise process which incorporated one of the above resonant transitions as an intermediate step. The relative ion yield was determined as a function of laser wavelength and the resulting data analyzed to obtain absorption profiles and line shifts for nd, ns → 3p transitions.

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B. We report the total Penning ionization cross sections for He(2^3S_1) atoms incident on Cd and Zn. The measurements are made for relative collision velocities above thermal by using a beam. Previous metal vapor studies of Cd and Zn have been confined to thermal velocities in pulsed afterglow experiments.

We also report the observation of HgBr(B-X) fluorescence resulting from collisions of $HgBr_2$ with a vibrationally excited beam of nitrogen molecules in the $A^3\Sigma_u^+$ electronic state.

TECHNICAL REPORT

I. Kinetic Processes in High Pressure Gases

II. Metastable Interactions
   1. Penning Ionization
   2. Dissociative Excitation

L. D. Schearer & R. H. McFarland

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Rolla, MO 65401

Technical Report to the Office of Naval Research
Grant No. N00014-75-C-0477 for the period December 1979 - December 1980.

CONTENTS

PERSONNEL

Co-Principal Investigators

Prof. Laird D. Schearer, project director
Prof. Robert H. McFarland*

Also associated with the program:

Visiting Assistant Prof. Javed Husain**
Mr. Daniel Krebs, Graduate Assistant
Mr. John Daniels, Graduate Assistant
Dr. David Fahey***

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CONTRACT DESCRIPTION

The purpose of the research program is to investigate energy exchange and energy loss mechanisms in discharge plasmas and their afterglows. Specific mechanisms currently under investigation in our laboratory are: (a) metastable interactions, such as dissociative excitation and Penning ionization, (b) collisional broadening and level shifts in excited states produced by the presence of a buffer gas and/or electrons, and (c) electron loss mechanisms due to volume recombination.

INTRODUCTION

Energy balance in an electrical discharge in a gas is determined by collisional-radiative processes involving excited atoms, ions, electrons, and buffer gas atoms. With the current interest in intense, high pressure discharges and their afterglows, the role of collisional-radiative processes involving highly excited (Rydberg) states has taken on a new importance. Such plasma processes as laser fusion, switches, and gas laser discharges have focused attention on the lack of adequate understanding of many of the collisional processes involving excited states.

For example, in a Na cell containing atom densities exceeding 10^{16} atom/cm^3 a modest laser power tuned to the 3s-3p transistion can totally ionize the vapor.[1] This is a highly efficient method of producing ionization in the vapor.[2] The energy is deposited in the system via an optical transition from the 3s to the 3p states. Subsequent collisional processes involving associative excitation and superelastic e-excited atom collisions redistribute this energy. The effects of collisions appear also as line broadening and level shifts which further influence energy redistribution effects.

We are investigating such energy-transfer and energy loss mechanisms which occur at high pressures and at relatively high densities of excited state species. In general it is difficult to obtain a uniform discharge at high buffer gas pressures (> atmosphere). Consequently, we utilize high power, fast-pulsed lasers to produce the required high densities of excited states and ion/electron concentrations. The excited state concentrations can be observed by monitoring optical emission from the various excited states. Time dependent processes are observed by examining the time dependent emission in the afterglow of fast, high-power resonant laser pulses applied to the system.

PROGRESS REPORT

A. Kinetic Processes in High Pressure Gases

B. Metastable Interactions

   1. Penning Ionization

   2. Dissociative Excitation

A. KINETIC PROCESSES IN HIGH PRESSURE MIXTURES OF Na-Ar

1. INTRODUCTION

Much work has been done in the area of laser production of plasmas by cascade ionization and multiphoton absorption.[3] Some experiments have utilized an intermediate resonance transition to enhance the degree of ionization achieved. Most such experiments have relied on associative or other collisional processes as intermediate steps in the ionization. Notably Lucartorto and McIlrath have achieved almost complete ionization of sodium ($n_{Na} \sim 10^{16}$ cm^{-3}) by exciting the D line transitions with the output from a flashlamp pumped dye laser.[4,5] Since the 3^2P states are considerably less than halfway to the ionization limit, those results generated much interest and much work has been done attempting to determine the ionization mechanisms.[6,7,8] Since the cross-section for two photon absorption from the excited 3^2P states is too small to account for the high degree of ionization observed,[8] the proposed mechanisms have involved collisional processes, or stimulated Raman absorbtion as an intermediate step.

In contrast to the experiments of Lucartorto and McIlrath, Lam[9] achieved a high degree of ionization in potassium but attributed most of the ionization to purely radiative processes. He found that by exciting the $4\ ^2S_{1/2} - 5\ ^2P_{3/2}$ transition in potassium with a 4 ns 120 uJ laser pulse, a dense ($n_e = 10^{14}$ cm^{-3}) plasma was produced. Lam suggested that the ionization mechanism was photoionization from the $5\ ^2P$ states.

In this experiment, a laser induced plasma was produced in a sodium-argon system. The first step of the excitation is less than halfway to the ionization limit. A second laser causes ionization; operating at a wavelength such that the electron is ejected with essentially thermal velocity. The initial conditions of the afterglow in this experiment are thus much different from those in Lam's experiment, and differences in the plasma produced were observed.

Since the effects of the two laser fields could be studied independently or together, it was possible to positively attribute most of the observed ionization to photoionization of the $3\ ^2P$ states. The fact that relatively little ionization was observed when only the D lines were excited might seem inconsistent with Lucartorto and McIlrath's findings. The principal difference between this experiment and theirs was the laser pulse length. The power levels and sodium densities were otherwise comparable.

APPARATUS

The reaction cell was stainless steel with sapphire windows pressed to the cell body inside the oven. Stainless steel side arms extended outside the oven to cooled pyrex windows which formed a vacuum seal. The cell was leak tested with a helium mass spectrometer leak tester and found leak tight at the highest sensitivity of the instrument. Two opposing windows allowed the laser light to pass through the reaction region with minimum scattering. A third window allowed monitoring the fluorescence 90° to the direction of the lasers. The fluorescence was detected by a S-20 photomultiplier with a fast voltage divider chain for enhanced linearity. The monochromator was a 1/2-meter Jarrel-Ash with 1190 groove/mm. grating. The signal was routed to either a TEC 5441 storage/sampling oscilloscope, a Par 160 boxcar integrator, or an SSR 1110 synchronous photon counter. The storage/sampling oscilloscope was used for recording fast transients which produced smooth displays of the observed wave forms. The boxcar integrator was used to measure intensities of the time resolved spectral lines. The photon counter was used for high resolution scans which measured the shift and broadening of the line emission. A standard sodium lamp provided wavelength calibration. A quartz tungsten standard lamp was used for absolute intensity calibration of the detection system.

The dye lasers each provided 5 ns, 65 μJ pulses at their respective wavelengths. The pump laser was a 500 KW NRG N_2 laser operating at 30 Hz. (See Fig. 1.)

EXPERIMENT

The cell was baked out to 500°C for 12h at a pressure of 10^{-6} Torr. Sodium was titrated into a reservoir attached to the reaction cell. Research grade argon was admitted to the desired pressure. In order to accurately determine the buffer density, a Wallace-Tiernan pressure gauge was left attached to the cell during operation. Duffision of sodium to the cool windows was negligible provided the buffer pressure was maintained above 100 Torr.

Figure 2 is a sampling/storage oscilloscope trace of the $4\ ^2D - 3\ ^2P$ fluorescence. The rise of the signal corresponded closely to the firing of the lasers. The initial decay was rapid ($\sim$ 100 ns) followed by a slower ($\sim$ 2 us) decay. The fast component signal was dependent only on the D line excitation in the 3d, 4d, 5d - 3p fluorescence. This observation is consistent with those of Allegrini, et al,[10] who excited the D lines of a dense sodium-neon mixture with a CW laser. They observed fluorescence from levels with close to twice the energy of the $3\ ^2P$ states and attributed the fluorescence to states formed in the process

$$Na(3P) + (Na(3P) \rightarrow Na(3S) + Na(Nx) \pm E$$

where E is the energy defect or excess to be transferred to kinetic energy.

The slower decay was due to excited states created in the recombination process. There was some slow component signal when only the D lines were excited but the slow component was sufficiently strong for plasma diagnostics only if additional excitation was employed. The additional excitation was accomplished by either tuning the blue laser to a high level resonance or by operating the blue laser within a few 100 cm^{-1} above the ionization limit.

For comparable laser power levels, excitation of the 7d level was approximately twice as efficient for ion production as laser excitation just above the ionization limit. The fraction of atoms ionized for the former case was estimated to be about 10% at 378°C ($n_{NA} \sim 3 \times 10^{15}$ cm^{-3}).

Figure 3 is a wavelength scan of the slow component fluorescence 1μs after the laser has fired. Excited state populations at different times in the afterglow were obtained from such scans by an absolute calibration of the detecting apparatus; e.g., $n^* = I_\nu/(AgDV)$ where D is the overall detection efficiency, V is the volume of the observed region, g is the degeneracy of observed states, I_ν is the observed intensity in photons/sec, and A is the transition probability. Because of uncertainties in estimating the volume of the observed fluorescence and the overall detection efficiency, the absolute intensities of these lines have a systematic uncertainty of about a factor of two. The relative intensities are considered accurate within at least 10% (the accuracy declines with member number because the S/N ratio decreases), and electron temperature data depends only on relative intensities. Furthermore, comparisons of relative electron densities at different times in the afterglow are possible since the effect of miscalibration is only a change in the scale of n_e^2.

DISCUSSION AND CONCLUSIONS

Figure 4 is a plot of excited state density versus energy level. In the region of local thermodynamic equilibrium, excited state density is given by Saha's equation:

$$\frac{n^*}{g^*} = \frac{ne^2}{2g_{ion}} \left(\frac{h^2}{2\pi mKTe}\right)^{3/2} \exp\left(\frac{(Eion-E^*)}{KT3}\right)$$

Thus, in this region, the plot of log (n^*/g^*) vs E^* is linear. The region of local thermodynamic equilibrium extends from the ionization limit to lower levels for which collisional excitation is more likely than radiative decay. Greim[11] has calculated that the level, n, for which radiative decay and collisional excitation are equally likely is given approximately by:

$$n' \simeq 1s6\, n_e^{-2/17} \frac{KTe}{E_H}^{1/17} e^{\frac{4\,E_H}{17n^{13}\,KT_e}}$$

For $n_e = 1 \times 10^{14}$ cm^{-3}, Te = 800°K, n ~ 4.

Departure from local thermodynamic equilibrium was observed to begin at n=8 indicating that collisional processes dominate radiative processes for $n \geq 8$.

Table 1 is a table of measured electron densities and temperatures for two methods of plasma production. It is interesting to note that although the degree of ionization is greater with the resonant excitation, the electron temperature is roughly equal in both cases. The temperatures are much lower than those measured by Lam in his experiment in which the electrons were ejected with considerable energy. Thus most of the electron energy in Lam's experiment would appear to come from the energy excess generated in the photoionization process.

Figure 5 is a scan of the 5 D - $3P_{3/2,1/2}$ recombination lines. The shift, broadening, and asymmetry of the line profile is evident. Because electrical discharges require lower buffer densities, previous observations of collision broadened atomic lines connecting excited states have been at lower perturber densities and have required an interferometer for resolution.[12,13] The shifts and broadenings observed exceeded those predicted by perturber broadening alone[12], particularly at high member number. Scans were taken at different delay times to determine if Stark broadening might contribute. Calculated Start shift parameters for sodium are on the order of those observed and increase with member number[11]. No variation in shift with delay time (and therefore electron density) was observed. The buffer pressure was varied and the shifts were found to be approximately linear in perturber density.

Table 1

Excitation	Time (μs)	Electron Density (cm^{-3})	Electron Temp (nK)
λ_1 = 589 nm (3s→3p)	1	1.4×10^{14}	1060
λ_2 = 403 nm (3p→Na+)	2.5	6.4×10^{13}	809
	5	5×10^{13}	850
λ_1 = 589 nm (3s→3p)	1	3.4×10^{14}	1060
λ_2 = 466.8 nm (3p→7d)	2	2.0×10^{14}	876
	4	1.3×10^{14}	853

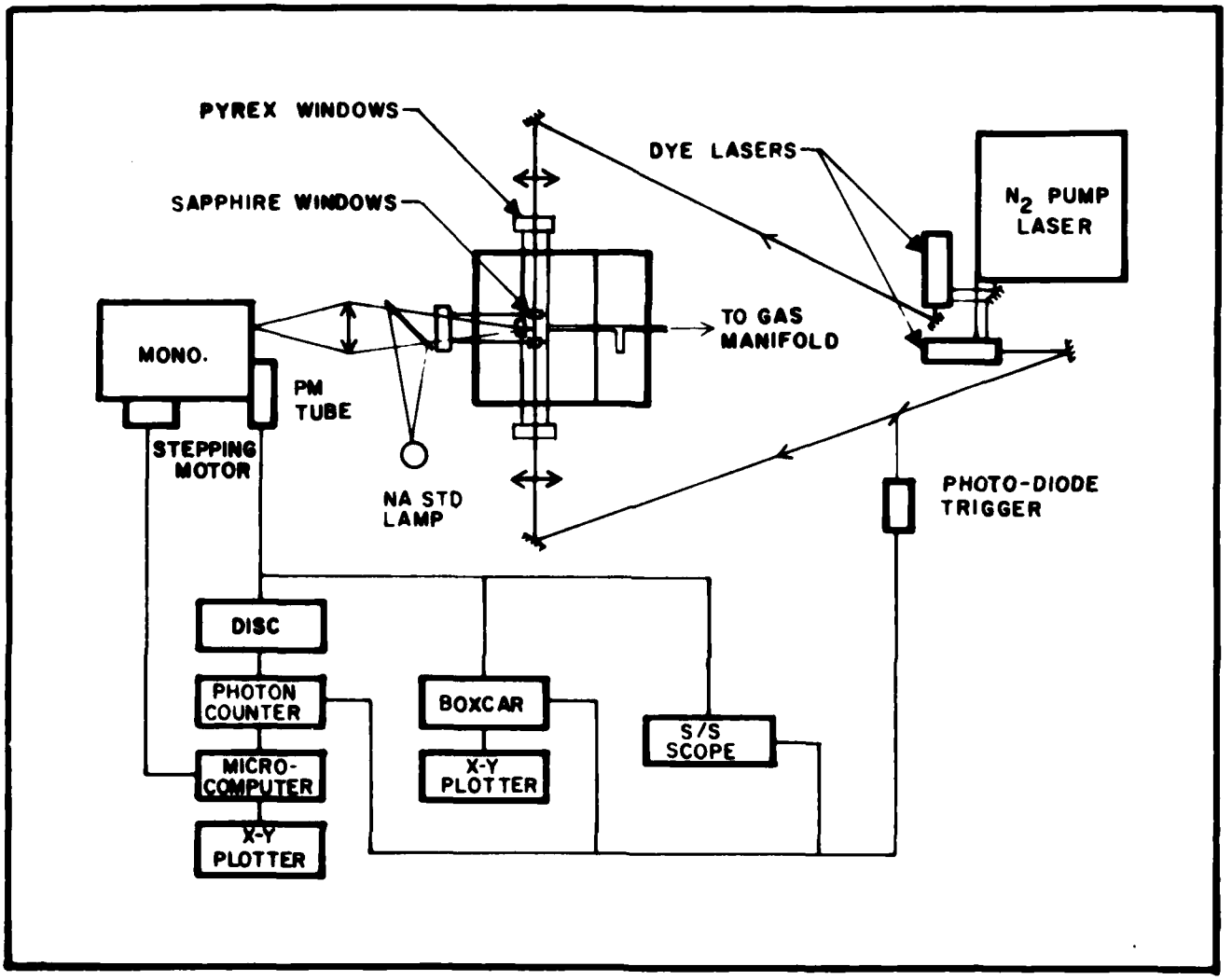

Figure 1. Schematic of the apparatus.

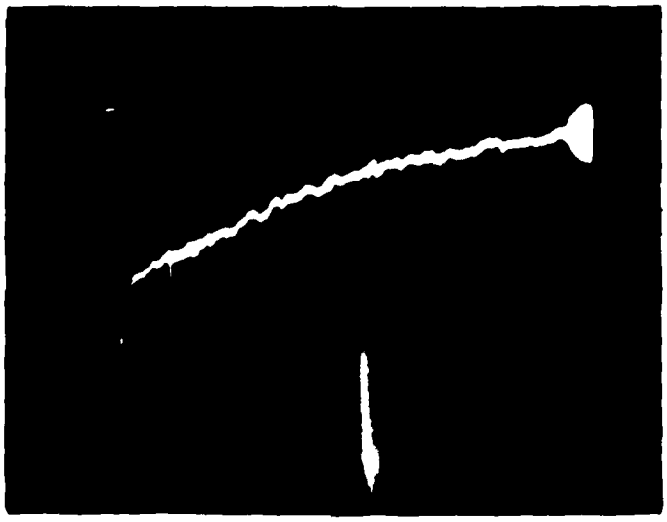

Figure 2. Sampling/storage oscilloscope trace of 4d-3p fluorescence. Slow decay tail is due to excited states formed from electron-ion recombination. Scale is .2 V/div vertical by .2 usec/div horizontal.

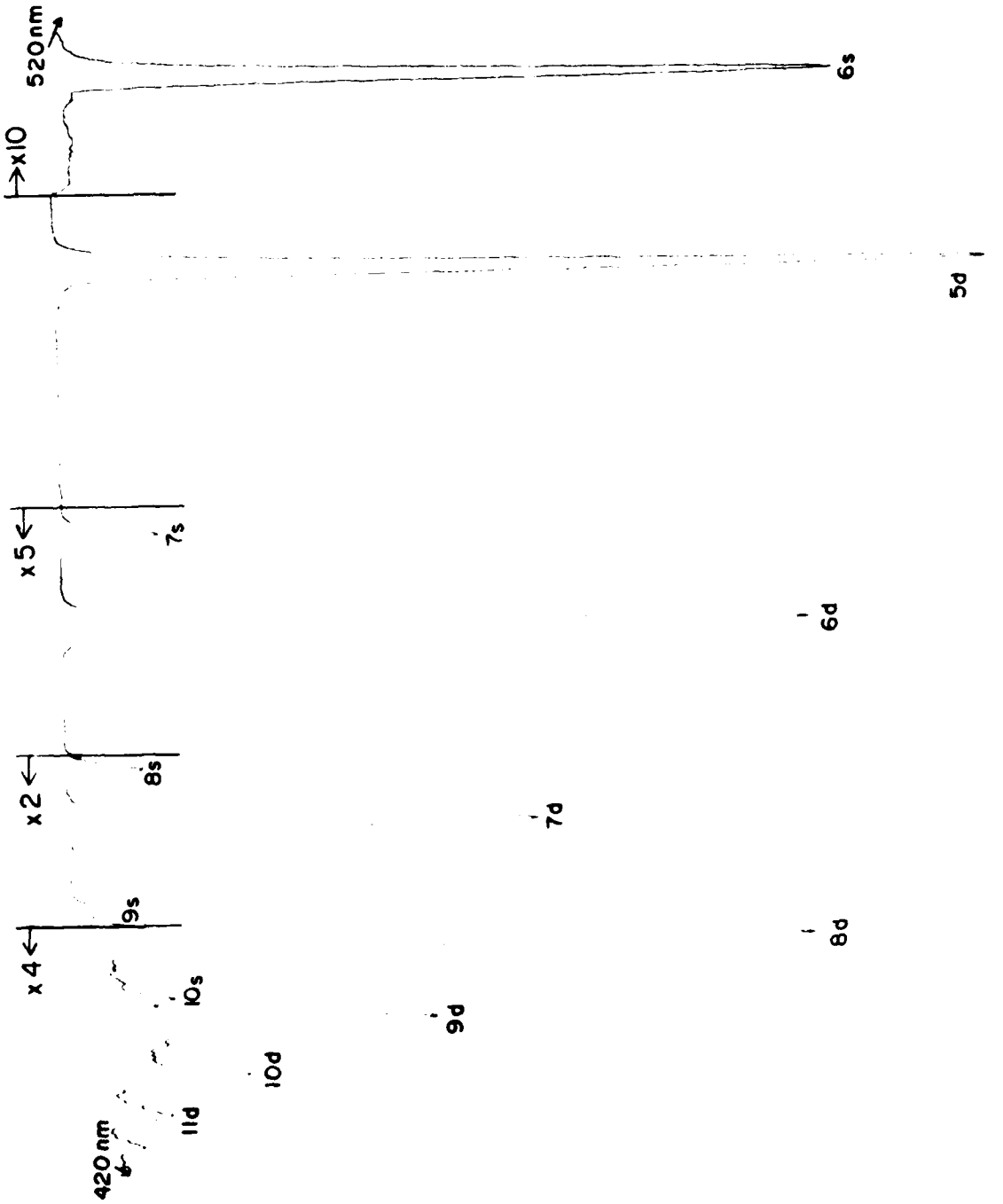

Figure 3. Wavelength scan of the recombination fluorescence. All lines are n -3p type transitions. Resolution is .8 nm (FWHM).

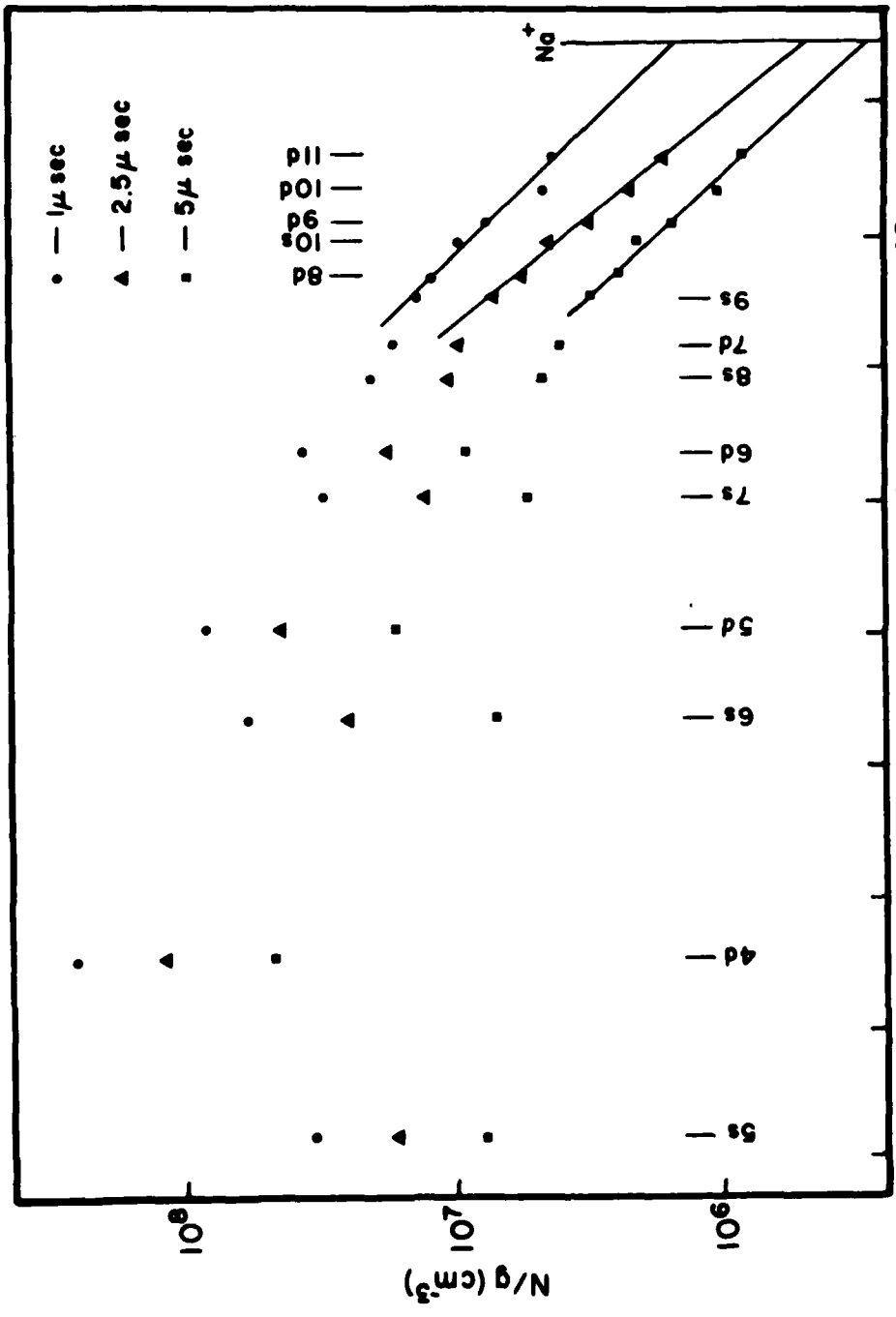

Figure 4. Density per quantum state versus excited state energy.

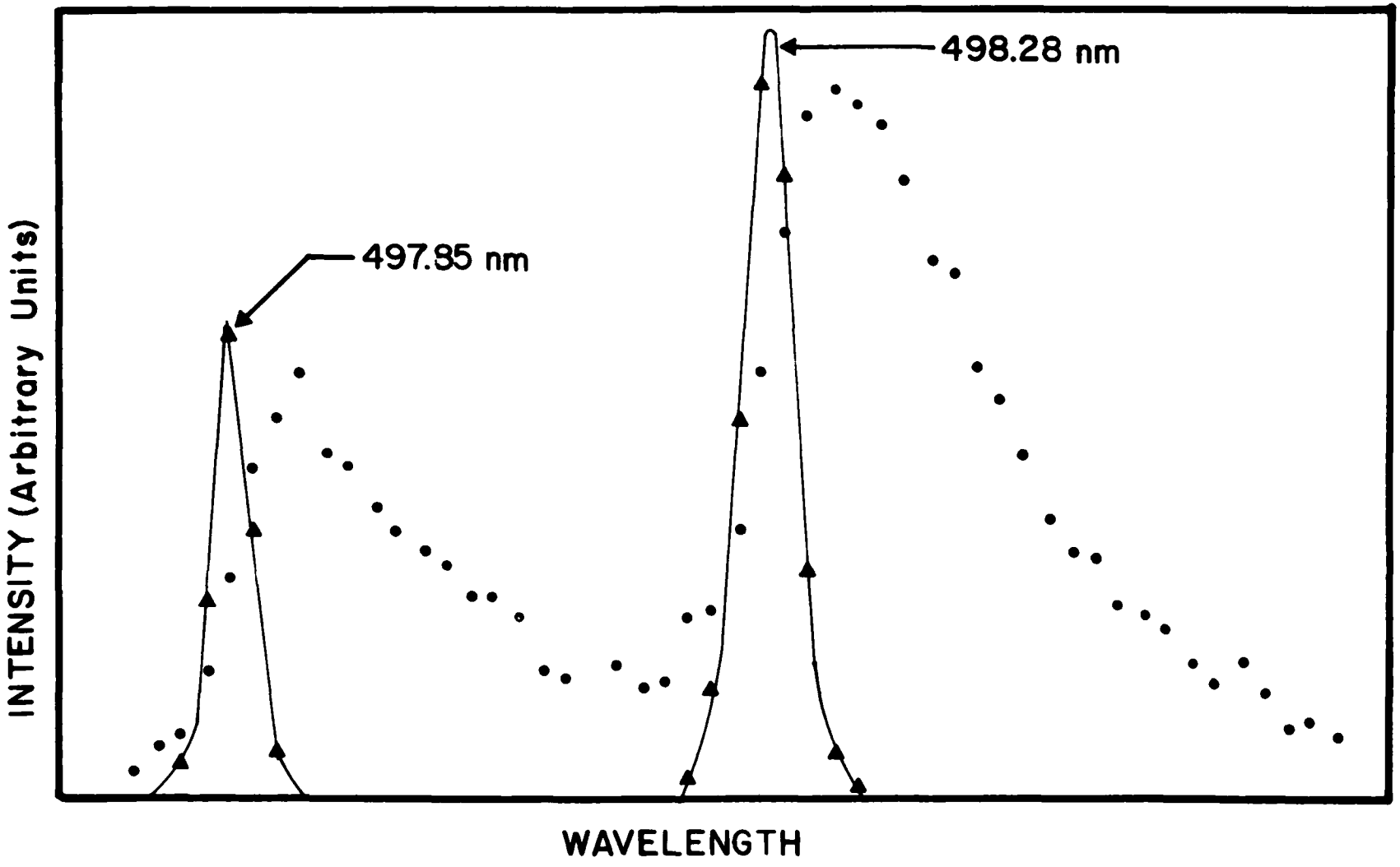

Figure 5. Wavelength scan of 5d-$3P_{3/2,1/2}$ recombination fluorescence. Scan was taken point-by-point with the photon counter recording counts from the plasma fluorescence (•) and the sodium lamp (▲) at each point. The lamp signal provides a wavelength calibration and a check on instrumental resolution.

B. METASTABLE INTERACTIONS

1. PENNING PROCESSES

The measurement of Penning ionization cross sections for collisions between rare-gas metastable atoms and metal vapors has been of recent interest following the development of metal vapor lasers. Of particular interest has been the group II metal vapors. Lasing action has been observed in the ion levels of Cd, Zn, Ca, and Sr when these elements are excited in He discharges.[1-4] The metal vapor excitation mechanism for most lasing lines has been shown to be Penning ionization by He metastable atoms through excitation cross-section measurements. Further, there is continuing interest in the dependence of these Penning cross sections on particular atomic parameters. A study of the variation of the cross section of Cd and Zn with rare-gas metastables found the results consistent with the fact that the cross sections are proportional to the cube of the "hard-sphere" radii of the collisions.[14]

We have determined the total Penning ionization cross sections for $He(2\ ^3S_1)$ atoms incident on Cd and Zn. The measurements are made for relative collision velocities above thermal by using a beam.[15] Previous metal vapor studies of Cd and Zn have been confined to thermal velocities in pulsed afterglow experiments.

2. DISSOCIATIVE EXCITATION

Dissociative collisions involving the mercuric halide compounds $HgBr_2$, $HgCl_2$, and HgI_2 have been of recent interest because of laser output achieved on the $B^2\Sigma$-$X^2\Sigma$ transition in the respective mercuric-halide radicals in the range of 400-600 nm. Population inversion has been achieved by photodissociation and electron impact excitation in mixtures of the mercuric-halide compounds and rare gases. Recently, Burnham has noted improved

laser efficiency and output power when nitrogen gas has been added to the lasing mixture.[16] The improved performance with added N_2 is likely due either to direct pumping of the upper laser level by excited states or to collisional dissociation of the ground state radical which prevents bottlenecking.

We have observed the HgBr(B-X) fluorescence resulting from collisions of $HgBr_2$ with a vibrationally excited beam of nitrogen molecules in the $A^3\Sigma_u^+$ electronic state. We have demonstrated that a vibrationally excited $N_2(A)$ beam dissociatively excites $HgBr_2$ producing in part $HgBr(B^2\Sigma^+)$.[17] We assume that all vibrational levels of sufficient energy participate in this excitation. Further, we infer that certain vibrational levels can dissociatively excite $HgBr_2$ producing $Hg(6^3P)$. We expect that the same study made with $HgCl_2$ and HgI_2 will yield similar results.

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High flux beam source of thermal rare-gas metastable atoms

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IEEE JOURNAL OF QUANTUM ELECTRONICS, VOL. QE-16, NO. 1, JANUARY 1980

Injection-Locked Dye Laser Pumped by a Xenon-Ion Laser

E. R. CARNEY, D. W. FAHEY, AND L. D. SCHEARER

Abstract—**Injection locking of a dye laser is reported for a 4-mirror ring-cavity dye laser pumped by a xenon-ion laser. Both a He–Ne laser and tunable CW dye laser were used as the injection sources.**

Volume 74A, number 6 PHYSICS LETTERS 10 December 1979

HANLE LIFETIME MEASUREMENTS OF SrI $^1P_1$ AND CaI $^1P_1$ LEVELS EXCITED BY A NEUTRAL BEAM OF 1 $^1S_0$ HELIUM ATOMS☆

D.W. FAHEY, W.F. PARKS and L.D. SCHEARER
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Received 25 July 1979

A 0.8 keV He(1 $^1S_0$) beam was used to coherently excite the $^1P_1$ levels of Sr and Ca targets. The coherence appears as an alignment of the excited state with respect to the beam axis. We report here the results of a Hanle measurement, or a zero-field level-crossing experiment, performed on these coherently excited levels. The radiative lifetimes of the SrI $^1P_1$ and CaI $^1P_1$ levels were measured to be 4.7 ns and 5.3 ns, respectively. These values are in good agreement with conventional Hanle measurements.

Dissociative excitation of $HgBr_2$ in collisions with a beam of metastable nitrogen[a)]

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(Received 29 January 1980; accepted 18 February 1980)

In this note we report the observation of $HgBr(B-X)$ fluorescence resulting from collisions of $HgBr_2$ with a vibrationally excited beam of nitrogen molecules in the $A\,^3\Sigma_u^+$ electronic state. The beam source used was a glow-discharge maintained in pure N_2 undergoing nozzle expansion.[9,10] The most probable energy of the resulting beam was 180 meV and a lower limit on the flux value was determined to be 10^{14} atoms/s sr.

J. Chem. Phys. 72(11) 1 June 1980

Total Penning ionization cross sections of Cd and Zn for $He(2\,^3S_1)$ atoms[a)]

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(Received 17 September 1979; accepted 5 November 1979)

The total Penning ionization cross sections for Cd and Zn for $He(2\,^3S_1)$ atoms have been measured. The method used was that of Penning ion collection from a vapor cell through which a $He(2\,^3S_1)$ beam was passed. The beam energy was 66 meV with a velocity spread of 45%. The measured cross sections were 106(± 22) and 35(± 7) Å^2 for the Cd and Zn targets, respectively.

J. Phys. B: Atom. Molec. Phys., Vol. 12, No. 20, 1979. Printed in Great Britain

LETTER TO THE EDITOR

The Hanle effect in Penning-excited ions†

D W Fahey, W F Parks and L D Schearer

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Received 6 August 1979

Abstract. A thermal beam of helium (2^3S_1) metastable atoms was used to excite coherently the $^2P_{3/2}$ levels of Ca, Sr and Ba in a Penning ionising collision. The coherent excitation of the ions appears as a linear polarisation of the optical emission from the excited ions. The degree of linear polarisation is 5·5, 3·5 and 0·5% for Ca, Sr and Ba, respectively, with the polarisation parallel to the beam direction. Hanle effect signals from the $^2P_{3/2}$ level of Sr were observed and the radiative decay rate measured.

170 TRANSACTIONS OF THE AMERICAN NUCLEAR SOCIETY
VOL. 34, 170 (1980)

NUCLEAR ARCHAEOMETRY — II

Sponsored by Isotopes and Radiation Division

1. Nuclear Archaeometry with Lasers: Effect of Laser Bandwidth on Sensitivity, *Robert H. McFarland, Javed Husain, Laird D. Schearer (Univ of Mo-Rolla)*

Recently[1,2] we introduced the use of lasers for nuclear archaeometry, geochronology, and related problems and have coined[2] the name laser isotope dating (LID) for this new technique. In this paper we study the effect of the laser bandwidth on LID sensitivity.

OTHER PRESENTATIONS: CONTRIBUTED

1. "Laser Isotope Dating (Lid)", (with R. H. McFarland and J. Husain), Bull. Am. Phys. Soc. 24, 1185, (1979), Dec., DEAP-Houston.

2. "Total Penning Ionization Cross-Sections of Cd and Zn for He(2^3S) Atoms", (with D. W. Fahey and W. F. Parks), Bull. Am. Phys. Soc. 24, 1177, (1979), Dec., DEAP-Houston.

3. "Excitation of Cd, Zn, and Sr by a Beam of Active Nitrogen", (with D. W. Fahey and W. F. Parks), Bull. Am. Phys. Soc. 25, 115, (1980), (32nd GEC) - Pittsburgh.

4. "Laser Isotope Dating: A New Technique of Radioisotope Dating Using Lasers", R. H. McFarland, Javed Husain, and L. D. Schearer, IVth International Conference in Nuclear Methods in Environmental and Energy Research, 14-18 April 1980.

5. "Absorption Profiles for Transitions to Sodium Rydberg Levels Perturbed by High Concentrations of Argon", D. Krebs and L. D. Schearer, 33rd GEC - Norman.

INVITED COLLOQUIA

Texas A&M University - December 1979

University of Texas - Dallas - December 1979

University of Arkansas - March 1980

University of Tennessee - April 1980

CONFERENCES ATTENDED

1. 33rd Annual Gaseous Electronics Conference

2. DEAP - Houston

3. IV International Conference on Nuclear Methods

4. Gordon Conference on Atomic and Molecular Interactions

5. 7th International Conference on Atomic Physics

OTHER FEDERAL SUPPORT

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